Scanning Tunneling Microscopy Characterization of the Electrical Properties of Wrinkles in Exfoliated Graphene Monolayers

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Supporting Information:

Figure S1. Process flow schematics for the fabrication of graphene sheets.

Figure S2. Hall measurement of graphene.

Figure S3. Locating the graphene sheet in STM.

Figure S4. Additional atomically-resolved STM topographs of the graphene samples investigated in this study.

Figure S5. Additional representative topographs of the observed wrinkles on graphene.

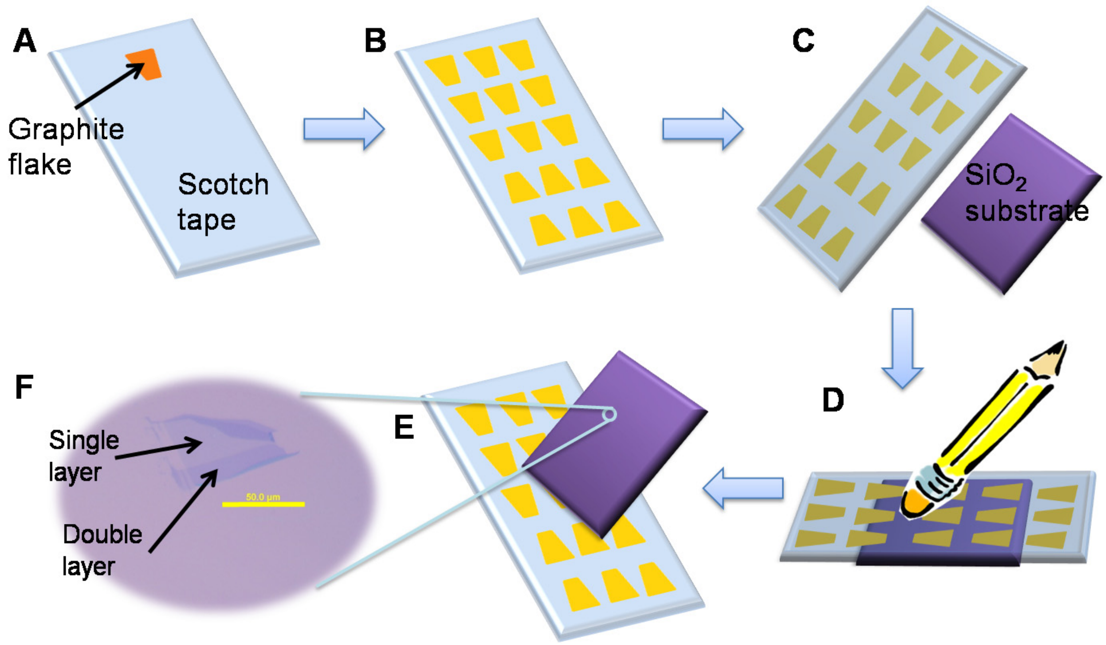

Figure S1. Process flow schematics for the fabrication of graphene sheets. (A): A thin Kish graphite flake is stuck onto a scotch tape. (B): By repeatedly folding and peeling the tape for ~10 times, the graphite flake is exfoliated into multiple thinner flakes, covering the entire tape surface. (C): The scotch tape is turned over, and the graphite flakes on the surface are brought into contact with a freshly cleaned SiO_2 substrate. (D): An eraser is used to gently rub the back of the tape, to ensure close contact between the graphite flakes and the substrate. (E): The scotch tape is peeled off from the substrate, leaving graphene sheets and other thin graphitic layers on the SiO_2 substrate. (F): Graphene sheets on the surface are identified through an optical microscope. Single-layer and double-layer parts of the graphene sheet (as confirmed through spatially resolved Raman spectroscopy) are labeled in graph. Scale bar: 50 μm.

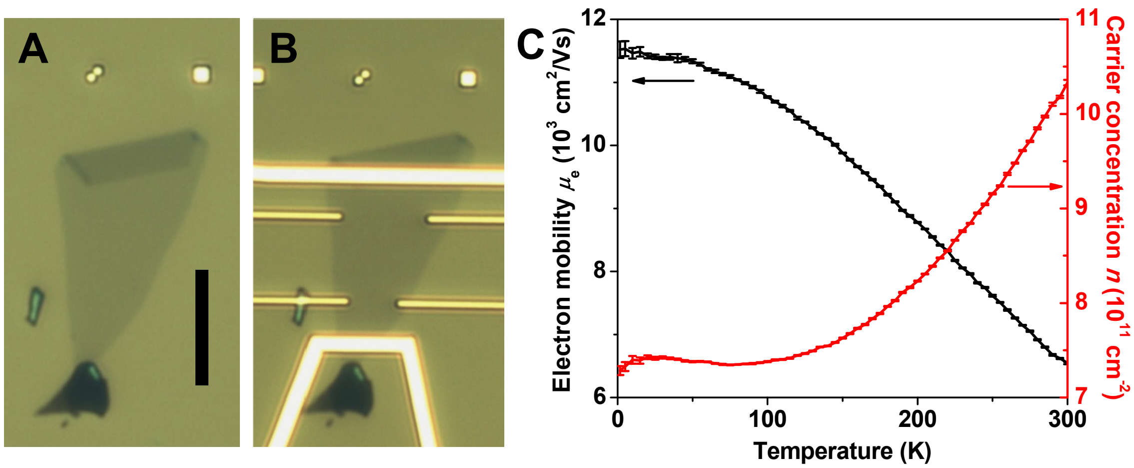

Figure S2. Hall measurement of graphene. (A): Optical microscope image of a graphene sheet (on 90 nm SiO_2 substrate) investigated in this study. Alignment markers were fabricated close to the graphene sheet using electron-beam lithography (EBL). Scale bar: 10 µm. **(B):** EBL was used to put down Ti/Au electrodes for Hall measurement. The electrodes also serve as guides for STM imaging (Fig. S3). **(C):** Electron mobility and carrier concentration obtained through Hall Effect measurement, as a function of temperature. Room temperature electron mobility is 6,500 cm^2/Vs.

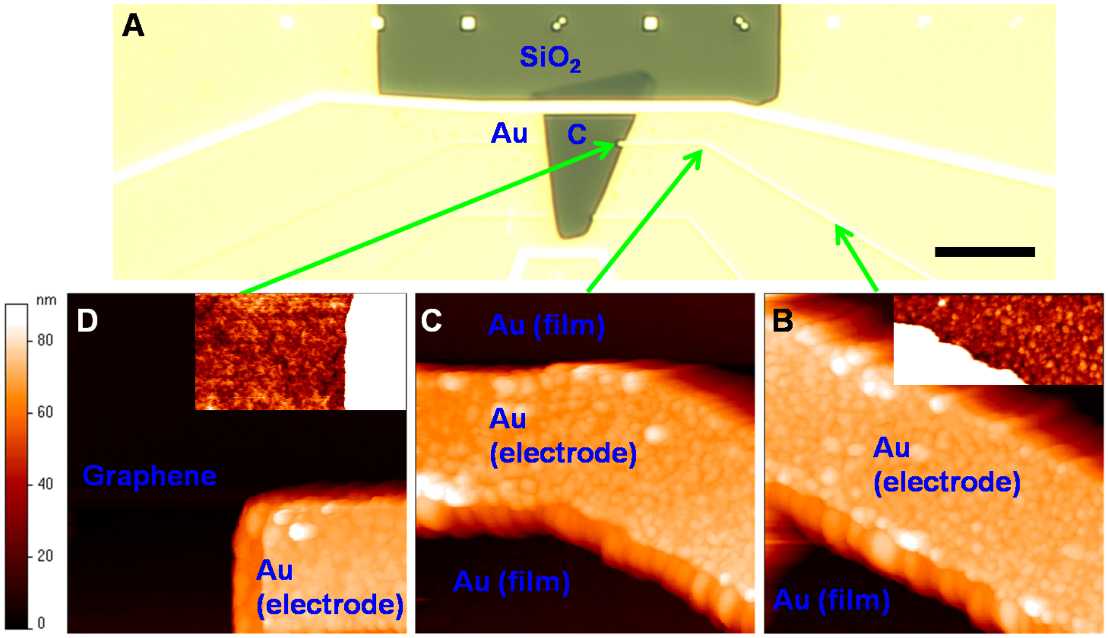

Figure S3. Locating the graphene sheet in STM. STM only images conducting surfaces. To prevent the STM tip from crashing into the sample, it's essential to avoid scanning over insulating surfaces, including the SiO_2 substrate used in this study. On the other hand, positioning of the STM tip under optical microscope has poor location control (~100 μm). This obstacle can be overcome by following the method described here. **(A):** An optical microscope image of a graphene device for STM study. Graphene is labeled as "C" for carbon. Scale bar: 10 μm. The graphene sheet is contacted at all edges with gold, so that the tunneling current diffuses in-plane through the gold film. Using optical microscope, the STM tip can be easily positioned on the conductive gold film (~500 μm × 500 μm). **(B)-(C):** STM topographs (800 nm × 800 nm) demonstrating how the graphene sheet is located for STM imaging. Large scale (~2 μm) scans are first performed to find the raised electrodes in the Au film. The graphene sheet is then located by tracing the electrodes. **(B):** Topograph of an electrode when the tip is far from graphene. Inset shows the topograph of the gold film near the electrode (white) with a 5 nm height scale. At this scale nanoscale gold islands are clearly observed. **(C):** By tracing along the electrode, the tip is moved closer to graphene, and the turn in the electrode unequivocally identifies the tip position on the gold film. **(D):** Topograph obtained at the end of the electrode, where the graphene sheet is reached [cf. (A)]. Inset shows the topograph of the graphene sheet near the electrode (white) on a small (2 nm) height scale: ripples in the graphene are observed.

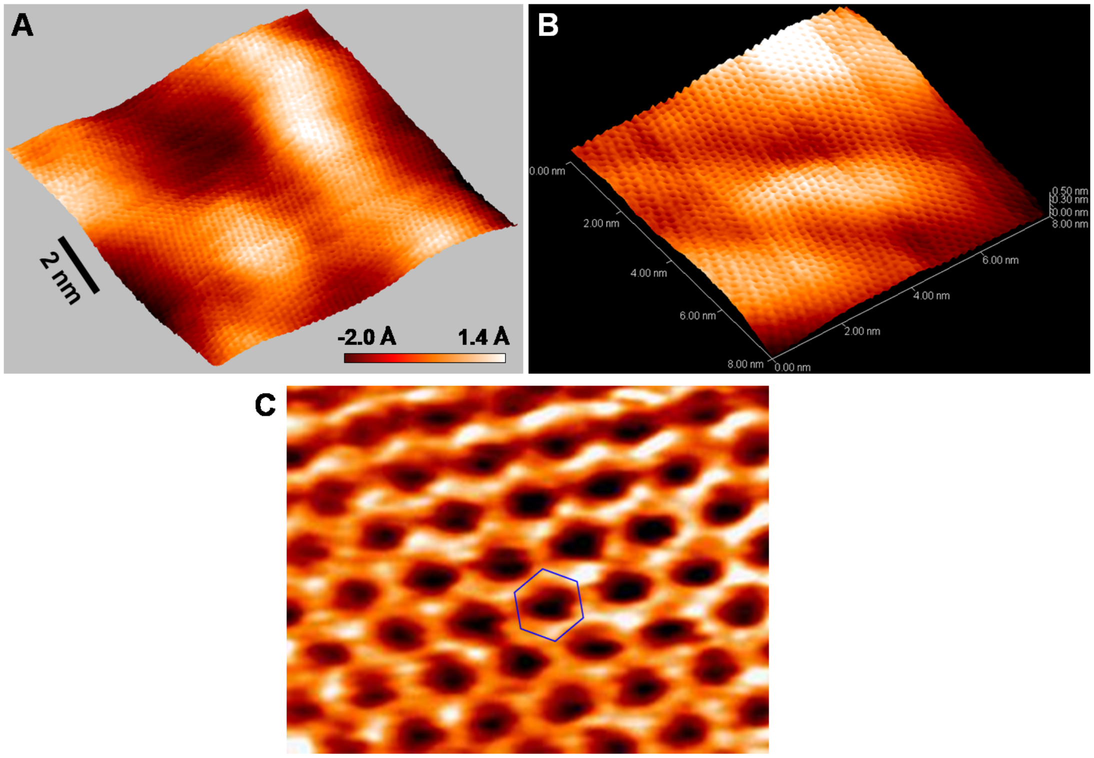

Figure S4. Additional atomically-resolved STM topographs of the graphene samples investigated in this study. (A): Constant-current STM topograph (V_b = 1.05 V, I = 290 pA) of a graphene sheet. (B): Constant-current STM topograph (V_b = 0.143 V, I = 124 pA) of another graphene sheet. (C): A close-up of the honeycomb lattice in (A). The blue hexagon has sides of 1.42 Å.

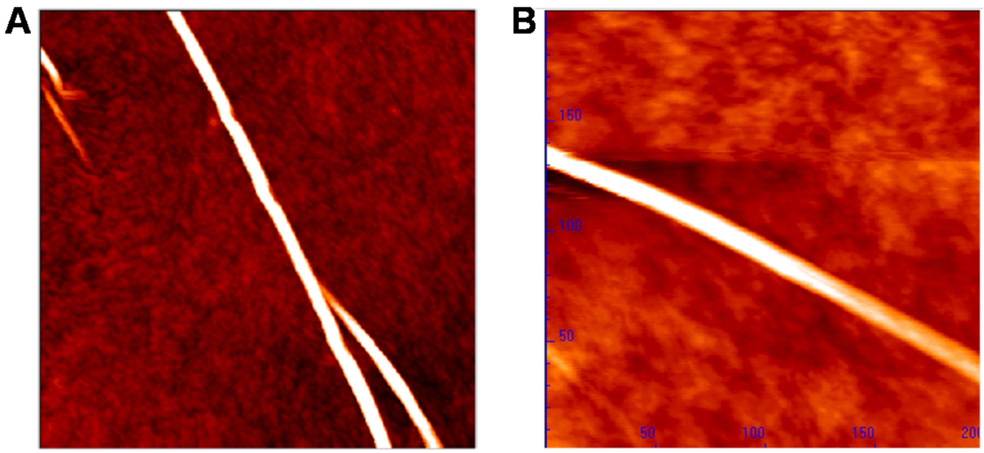

Figure S5. Additional representative topographs of the observed wrinkles on graphene. (A): An 800 nm × 800 nm topograph showing multiple wrinkles. The largest height in this topograph is ~5 nm. (B): A 200 nm × 200 nm topograph showing a wrinkle on another graphene sample. The largest height in this topograph is ~4 nm.